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Comparing mercury concentrations across a thirty year time span in anadromous and non-anadromous Arctic charr from Labrador, Canada

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HIGHLIGHTS

- Total mercury concentrations ([THg]) were measured in Arctic charr from Labrador.
- Wide-scale changes in age- and length-adjusted [THg] did not occur between 1977–78 and 2007–09.
- The mean age and fork-length of captured fish declined between the two time periods.
- Trends in [THg], age, and fork-length were similar in anadromous and non-anadromous Arctic charr.
- Significant warming trends were observed at Labrador weather stations between 1977–78 and 2007–09.

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ABSTRACT

Anadromous and non-anadromous Arctic charr (*Salvelinus alpinus*) from multiple sample sites in Labrador, Canada were used to investigate possible differences in total mercury concentration ([THg]) between 1977–78 and 2007–09. The mean [THg] of anadromous Arctic charr was 0.03 μg/g wet weight (ww) in 1977–78 and 0.04 μg/g ww in 2007–09, while mean concentrations in non-anadromous conspecifics were 0.18 μg/g ww in 1977–78 and 0.14 μg/g ww in 2007–09. After correcting for the effects of fish age and fork-length, there was no widespread difference in the mean [THg] of anadromous or non-anadromous fish between the two time periods. However, at individual sites sampled during both time periods, [THg] increased, decreased, or did not change. The mean age of sampled fish declined from 9.0 years in 1977–78 to 8.2 years in 2007–09 for anadromous fish, and from 11.7 years to 10.5 years in non-anadromous Arctic charr. Similarly, mean fork-lengths decreased from 450 mm to 417 mm in anadromous and from 402 mm to 335 mm in non-anadromous fish between 1977–78 and 2007–09. The mean annual temperature at four Labrador weather stations increased by 1.6 °C to 2.9 °C between the two sampling periods. The lack of an overall trend in anadromous or non-anadromous Arctic charr [THg] despite warming temperatures that favour increased mercury methylation suggests that regional changes in climate-driven factors have had limited impacts on mercury exposure in Labrador freshwater or marine fish.

1. Introduction

Mercury is a potent neurotoxin, and elevated concentrations in Arctic biota present an ongoing threat to the health of northern people (Stow et al., 2011) and wildlife (Dietz et al., 2011). The primary route of mercury exposure is through diet. Therefore mercury contamination is particularly concerning in species such as Arctic charr (*Salvelinus alpinus*), which are widely consumed by northern people and are culturally and economically important (Evans et al., 2005; Van Oostdam et al., 2005). Total mercury concentrations ([THg]) in Arctic charr vary widely, both within and among populations (Evans et al., 2005; Lockhart et al., 2005), and have been positively related to fish age, size, and trophic position (e.g., Gantner et al., 2009, 2010; Rigét et al., 2000; Swanson et al., 2011; van der Velden et al., 2012, 2013a). Life-history strategy is also an important determinant of Arctic charr [THg]. Anadromous Arctic charr, which feed in the marine environment, consistently yield lower mercury concentrations than non-anadromous conspecifics, which feed strictly in freshwater (Bruce and Spencer, 1979; Evans et al., 2005; Lockhart et al., 2005; Rigét et al., 2000; Swanson et al., 2011; van der Velden et al., 2012, 2013a). The lower [THg] in anadromous individuals may be related to differing ages, growth rates, or muscle C:N ratios between the two life-history types (Swanson et al., 2011), and/or may reflect lower prey [THg] due to lower mercury uptake at the base of marine relative to freshwater foodwebs (van der Velden et al., 2013b).

Recent climate warming trends, especially prominent at high latitudes (e.g., Walsh et al., 2011), have the potential to impact Arctic

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charr [THg] through alterations in population size- and age-distributions, and individual somatic growth rates (Reist et al., 2006). The prevalence of anadromy in Arctic charr is also likely to be affected, through changes in river flows and the timing of ice break-up, as well as increases in lacustrine and marine primary productivity (Finstad and Hein, 2012). Climate-related changes may also affect fish [THg] through enhanced mercury availability and cycling in aquatic food webs (Carrie et al., 2010), with increased river flows, coastal erosion, and permafrost degradation likely to deliver greater mercury inputs to aquatic systems (Stern et al., 2011). Furthermore, mercury methylation and uptake by aquatic biota may be amplified following increases in water temperature, nutrient availability, and productivity (Chételat and Amyot, 2009; St. Louis et al., 2005; Ullrich et al., 2001).

Mercury concentrations in many Arctic species have increased tenfold since pre-industrial times (Dietz et al., 2009). While recent time series show no consistent pattern of increasing or decreasing [THg] in biota throughout the circumpolar Arctic, increasing trends are apparent in some marine biota and freshwater fish, particularly from Canada and Greenland (Rigét et al., 2011). For landlocked Arctic charr, patterns of increasing muscle [THg] have been found in the Faroe Islands between 2000 and 2007 (Braune et al., 2011), and in southwestern Greenland between 1994 and 2008 (Rigét et al., 2010). Time trends for Arctic charr from the Canadian Arctic have varied spatially in significance and direction over a similar period. For example, [THg] in landlocked Arctic charr from Lake Hazen decreased for small (insectivorous) fish and showed no trend in large (piscivorous) fish from the early 1990s to 2009 (Muir et al., 2010). Muscle [THg] in anadromous Arctic charr increased at Cambridge Bay between 1992 and 2011, while there were no significant trends at Pond Inlet or Nain Bay (Evans and Muir, 2010, 2012).

Broad intraregional comparisons of temporal [THg] trends in anadromous and non-anadromous Arctic charr have not been conducted, and there is a paucity of temporal comparisons for anadromous populations from sub-Arctic regions where Arctic charr are heavily utilized (Dempson et al., 2008). Therefore, we investigated [THg] in multiple Arctic charr populations from Labrador, Canada, with a three-decade gap between sampling events (1977–78 and 2007–09). The objectives of the study were: [1] to assess whether there have been wide-scale temporal changes in anadromous and non-anadromous Arctic charr [THg] in Labrador, [2] to evaluate the influence of any changes in the mean size and age of sampled fish on [THg], [3] to determine whether temporal patterns in [THg] are consistent between anadromous and non-anadromous Arctic charr, and [4] to determine whether changes in Arctic charr [THg] are associated with variable environmental conditions, particularly an increase in mean annual temperature from the 1970s to present.

2. Methods

In the summers of 1977 and 1978, Fisheries and Oceans Canada (then the Department of Fisheries and the Environment) undertook wide-scale sampling of anadromous and resident freshwater fish in Labrador, Canada (Bruce et al., 1979). A total of 373 anadromous Arctic charr were obtained from 15 sample sites, and 146 non-anadromous Arctic charr were obtained from an additional 12 locations (Tables 1 and 2; Fig. 1). Subsequent sampling was conducted between 2007 and 2009 under the auspices of the International Polar Year scientific program, when 151 anadromous Arctic charr were obtained from 8 sample sites, and 113 non-anadromous Arctic charr were collected from an additional 5 locations (Tables 1 and 2; Fig. 1). Anadromous Arctic charr were obtained during both the early (1977–78) and recent (2007–09) time periods at four common sample sites: Hebron Fiord, Okak Bay, Tikkoatokak Bay, and Voisey Bay, while non-anadromous Arctic charr were collected from two common sites: Esker Lake and Tasialuk Lake, during both time periods. None of the sample sites were located within the Churchill River system, where the creation of the Smallwood Reservoir in 1971 and 1973 is known to have caused long-lasting elevated mercury concentrations in fish of the reservoir and downstream (Anderson, 2011).

During 1977–78, fish sampling was conducted using experimental gillnets of varying mesh sizes (mesh sizes not given), fyke nets, or by angling (Bruce et al., 1979). In 2007–09, non-anadromous Arctic charr were collected from lakes using multi-mesh nylon monofilament sinking gillnets (10–25 mm × 120 m and 10–60 mm × 120 m), and anadromous fish were collected from the nearshore marine environment using 114 and 127 mm mesh gillnets or by angling. Each specimen was measured for fork-length (±1 mm) and total weight (±0.01 kg), and fish ages were determined from sagittal otoliths using standard ageing methods (i.e., visual inspection of whole otoliths under reflected light, break and burn, and/or thin-section techniques). A sample of skinless dorsal muscle tissue was removed from the left side of each fish, and stored frozen prior to use in mercury analysis. Total mercury concentrations for the 1977–78 samples were measured via cold vapour atomic absorption spectrophotometry, using methods described in Uthe et al. (1970). Concentrations in the 2007–09 samples were measured via thermal decomposition, amalgamation, and atomic absorption spectrophotometry using a Milestone Direct Mercury Analyzer, DMA-80 (Milestone S.r.l., Sorisole, Italy) following U.S. EPA method 7473 (U.S. Environmental Protection Agency, 2007). The two methods have proven to yield comparable results for total mercury concentrations in fish muscle tissue (Butala et al., 2006). Quality control measures included the analysis of standard reference materials, as well as samples run in duplicate or triplicate, during each sample batch. The [THg] results are expressed as μg/g (parts per million) on a wet weight (ww) basis.

Meteorological data were obtained from the Environment Canada National Climate Data and Information Archive (www.climate.weatheroffice.gc.ca). Four Labrador weather stations were selected on the basis of proximity to Arctic charr sample sites and availability of monthly temperature data from 1970 to 2010: Nain A (56°33′ N, 61°41′ W), Hopedale (55°27′ N, 60°13′ W), Cartwright (53°42′ N, 57°02′ W), and Goose A (53°19′ N, 60°25′ W). Mean monthly temperatures were averaged to obtain annual mean temperatures, omitting years with any missing data.

All statistical analyses were performed using the R program for statistical computing (R Development Core Team, 2009) with Type I error set to $\alpha = 0.05$. Where mercury concentrations in the 1977–78 data set were below the limit of detection (<0.01 μg/g; $n = 12$ individuals, all from Hebron Fiord), half of the detection limit was used. Prior to inclusion in statistical analyses, mercury concentration data were natural-log (*ln*) transformed to obtain an approximately normal data distribution and remove heteroscedasticity. Means of two groups were compared using t-tests adjusted for variance equality or inequality, or using the nonparametric Wilcoxon rank sum test (Zar, 2010). The *ln* [THg] versus age and *ln* [THg] versus length relationships were investigated using Pearson's correlation and linear regression, with multiple models compared using ANCOVA. Linear mixed effects models were used to evaluate the effect of time period (1977–78 or 2007–09) or life-history type (anadromous or non-anadromous) on fish mercury concentration, age, and fork-length among sample sites. Mixed effects models were further used to explain variation in individual *ln* [THg] using time period, fish age, fork-length, or life-history type. Sample site was included as a random effect in all mixed models. Compliance with model assumptions was verified using diagnostic plots (e.g., fitted versus residuals, normal Q-Q plots of residuals and estimated random effects; Pinheiro and Bates, 2000; Zar, 2010).

3. Results

3.1. Anadromous Arctic charr

Individual muscle mercury concentrations ranged from <0.01 to 0.15 μg/g ww in the early time period (1977–78), and from 0.01 to

Table 1
Summary data (mean ± standard deviation) for anadromous Arctic charr collected from Labrador in 1977–78 and 2007–09.

Sample site	Location	Year	n	THg (μg/g ww)	Age (years)	Fork-length (mm)
Anaktalik Lake	56°30′ N, 62°50′ W	1978	3	0.05 ± 0.00	8.0 ± 0.0	520 ± 31
Double Mer Lake	54°02′ N, 59°35′ W	1977	12	0.04 ± 0.02	6.8 ± 2.1	264 ± 91
Hebron Fiord[a]	58°08′ N, 63°00′ W	1977	50	0.02 ± 0.01	10.2 ± 3.2	489 ± 107
Hopedale Bay	55°28′ N, 60°12′ W	1978	25	0.06 ± 0.03	9.1 ± 2.1	521 ± 57
Ikkudliayuk Fiord	60°12′ N, 64°31′ W	1978	67	0.02 ± 0.01	10.9 ± 1.4	503 ± 62
Makkovik Bay	55°10′ N, 59°00′ W	1978	18	0.05 ± 0.02	8.4 ± 1.1	496 ± 51
Nachvak Lake	59°00′ N, 64°08′ W	1978	21	0.01 ± <0.01	10.8 ± 1.5	495 ± 49
Napaktok Bay	57°58′ N, 62°35′ W	1978	8	0.03 ± <0.01	11.6 ± 3.3	469 ± 45
Okak Bay[a]	57°29′ N, 62°03′ W	1978	25	0.04 ± 0.02	8.8 ± 1.6	458 ± 51
Pack's Harbour	53°51′ N, 57°00′ W	1978	8	0.08 ± 0.02	6.9 ± 0.6	405 ± 11
Ramah Bay	58°52′ N, 63°12′ W	1978	25	0.02 ± 0.01	9.4 ± 1.9	454 ± 82
Sand Hill River	53°36′ N, 56°29′ W	1977	11	0.05 ± 0.02	8.2 ± 2.0	421 ± 72
Tikkoatokak Bay[a]	56°42′ N, 62°13′ W	1978	25	0.05 ± 0.02	8.0 ± 1.2	510 ± 59
Umiakovik Lake	57 24′ N, 62°50′ W	1978	50	0.03 ± 0.02	10.0 ± 2.9	476 ± 101
Voisey Bay[a]	56°15′ N, 61°54′ W	1978	25	0.05 ± 0.01	7.8 ± 0.9	518 ± 49
Anaktalak Bay	56°27′34″ N, 62°13′9″ W	2007	20	0.05 ± 0.01	9.4 ± 2.9	404 ± 58
Hebron Fiord[a]	58°5′38″ N, 63°2′27″ W	2009	21	0.03 ± 0.01	10.0 ± 3.5	498 ± 134
Nain Bay	56°37′44″ N, 62°31′12″ W	2008	20	0.02 ± 0.01	7.3 ± 2.0	363 ± 88
Okak Bay[a]	57°33′15″ N, 62°5′54″ W	2008	20	0.05 ± 0.03	7.9 ± 2.4	430 ± 98
Saglek Bay	58°32′41″ N, 63°27′30″ W	2007	22	0.02 ± 0.01	8.2 ± 3.6	389 ± 94
Throat Bay	56°17′54″ N, 61°47′26″ W	2007	17	0.05 ± 0.01	6.2 ± 2.4	258 ± 84
Tikkoatokak Bay[a]	56°45′41″ N, 62°29′56″ W	2007	20	0.04 ± 0.02	7.4 ± 2.7	428 ± 54
Voisey Bay[a]	56°17′9″ N, 62°4′29″ W	2007	11	0.07 ± 0.03	7.2 ± 1.9	499 ± 36

[a] Sampled during both the early and recent time periods.

0.14 μg/g ww in the recent period (2007–09), with site means presented in Table 1. The linear mixed model used to explain *ln* [THg] as a function of time period, given the random effect of sample site, produced mean *ln* [THg] estimates of −3.49 ± 0.14 (mean ± standard error) or 0.03 μg/g ww for Arctic charr from the 1977–78 period, and −3.30 ± 0.16 or 0.04 μg/g ww for Arctic charr from the 2007–09 period, with no significant effect of time period on *ln* [THg] ($F_{1,3} = 1.38$, $p = 0.33$). When the model was estimated using fish age and fork-length as covariates, there was a significant effect of age (linear mixed model; $F_{1,457} = 23.4$, $p < 0.001$) but no effect of length ($F_{1,457} = 0.01$, $p = 0.91$) or sampling period ($F_{1,3} = 3.1$, $p = 0.18$) on anadromous Arctic charr mercury concentration.

Fish ages ranged from 5 to 21 years in 1977–78 and 3 to 18 years in 2007–09, with site means presented in Table 1. A linear mixed model yielded mean age estimates of 9.0 ± 0.3 years (mean ± standard error) for the early time period and 8.2 ± 0.3 years for fish captured in the recent time period, and there was a significant decline in the mean age of captured anadromous Arctic charr from 1977–78 to 2007–09 ($F_{1,462} = 5.11$, $p = 0.02$). Measured fork-lengths ranged from 164 to 705 mm in the early sampling period and from 156 to 682 mm in the recent sampling period. A linear mixed effects model estimated fork-lengths of 450 ± 17 mm for the early time period and 417 ± 11 mm for the recent time period. As with age, there was a significant reduction in the mean fork-length of sampled anadromous Arctic charr from 1977–78 to 2007–09 ($F_{1,503} = 8.42$, $p < 0.01$). Within a given sampling site and year, there was a significant positive correlation between *ln* [THg] and age in 9 of 17 samples (53%) where sample size was ≥10, while significant positive correlations between *ln* [THg] and fork-length were evident in only 4 of 20 data sets (20%) with a sample size ≥10 (Table 3). In most cases, the correlation between mercury

Table 2
Summary data (mean ± standard deviation) for non-anadromous Arctic charr collected from Labrador in 1977–78 and 2007–08.

Sample site	Location	Year	n	THg (μg/g ww)	Age (years)	Fork-length (mm)
Cabot Lake	56°09′ N, 62°38′ W	1978	1	0.07	7	350
Esker Lake[a]	57°09′ N, 62°55′ W	1978	26	0.33 ± 0.15	15.0 ± 4.9	471 ± 108
Hebron (Ikarut) Lake	58°08′ N, 63°38′ W	1978	10	0.19 ± 0.12	13.8 ± 3.2	420 ± 45
Komaktorvik Lake	59°09′ N, 64°14′ W	1978	6	0.11 ± 0.07	14.5 ± 2.6	449 ± 29
Mistastin Lake	55°54′ N, 63°16′ W	1978	4	0.11 ± 0.08	10.3 ± 2.4	498 ± 46
Mistinippi Lake	54°47′ N, 61°19′ W	1978	13	0.50 ± 0.24	10.7 ± 2.2	405 ± 60
Nipishish Lake	54°10′ N, 60°46′ W	1977	7	0.35 ± 0.07	8.0 ± 0.8	301 ± 54
Saglek Lake[b]	58°49′ N, 63°21′ W	1978	17	0.19 ± 0.07	18.9 ± 4.0	491 ± 55
Shapio Lake	54°58′ N, 61°17′ W	1977	30	0.45 ± 0.44	8.9 ± 3.9	371 ± 173
St. Paul River	52°18′ N, 58°19′ W	1977	2	0.14 ± 0.02	6.5 ± 0.7	178 ± 4
Shallow Lake	57°39′ N, 63°16′ W	1978	6	0.32 ± 0.06	12.5 ± 1.6	432 ± 21
Tasialuk Lake[a]	56°45′ N, 62°45′ W	1978	24	0.09 ± 0.03	10.4 ± 2.2	354 ± 38
Coady's Pond #2[b]	56°38′30″ N, 63°37′30″ W	2007	20	0.12 ± 0.04	5.6 ± 3.0	316 ± 124
Esker Lake[a]	57°9′14″ N, 62°52′39″ W	2008	24	0.16 ± 0.23	12.0 ± 6.4	301 ± 110
Hebron Lake #2[b]	57°58′13″ N, 64°1′5″ W	2008	25	0.19 ± 0.17	12.2 ± 4.7	340 ± 82
Tasialuk Lake[a]	56°44′40″ N, 62°41′37″ W	2007	24	0.20 ± 0.04	11.1 ± 2.6	416 ± 41
Upper Nakvak Lake[b]	58°39′46″ N, 63°18′59″ W	2007	20	0.13 ± 0.12	11.7 ± 6.7	287 ± 166

[a] Sampled during both the early and recent time periods.
[b] Unofficial names. Coady's Pond #2 is a location name given to a lake that was sampled based on the confirmed presence of Arctic charr by Larry Coady during his exploration of this section of Labrador (Coady, 2008).

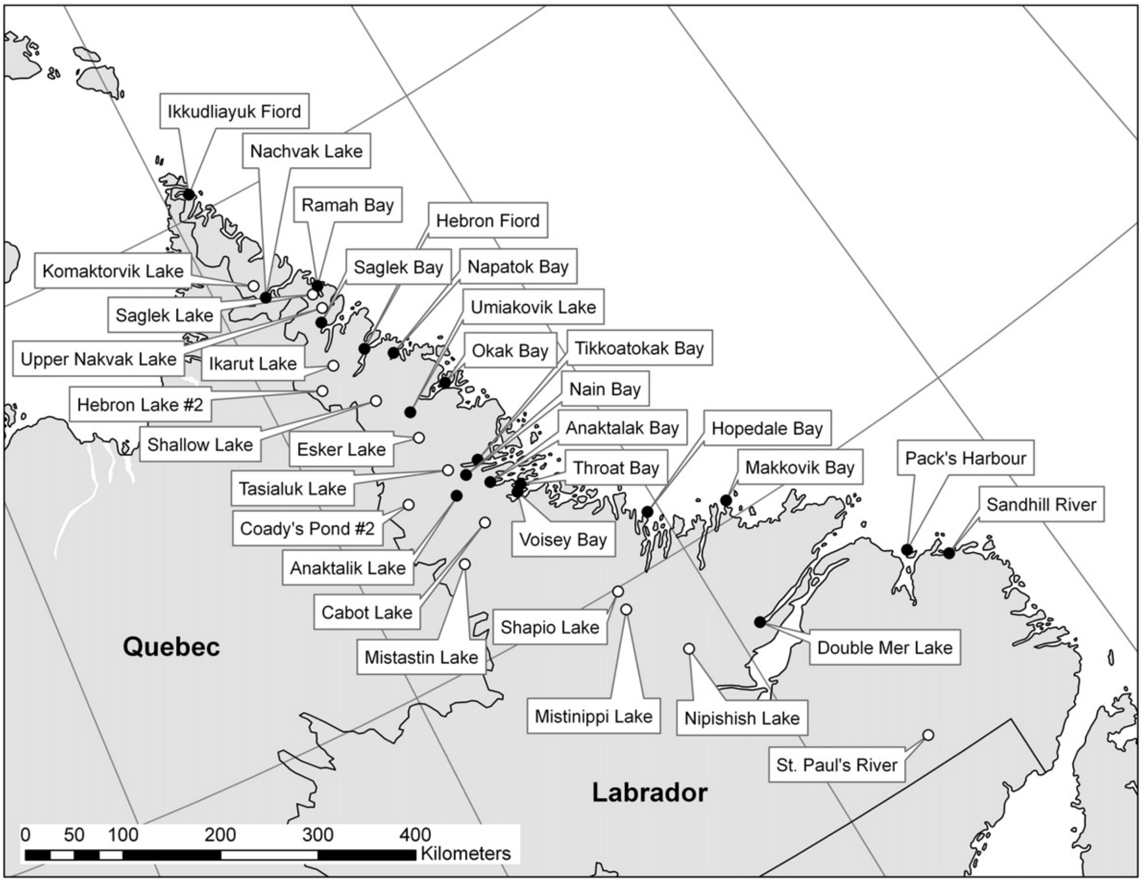

Fig. 1. Map of Labrador, Canada indicating the sampling locations of anadromous (black circles) and non-anadromous (white circles) Arctic charr.

concentration and fish age was stronger than the correlation with fork-length (Table 3). Age and fork-lengths were significantly correlated in 15 of 17 samples with a sample size ≥10 (Table 3).

Hebron Fiord was sampled in both 1977 ($n = 50$) and 2009 ($n = 21$). Age and fork-length distributions were similar in the two time periods, but mercury concentrations in the recent sample significantly exceeded those in the early sample (Table 4). Significant relationships between *ln* [THg] and age or fork-length were present only in the 2009 sample (Table 3; Fig. 2A).

Arctic charr were collected from Okak Bay in 1978 ($n = 25$) and 2008 ($n = 20$). There were no significant differences in the mean or variance of the sample age, fork-length or *ln* [THg] distributions between the two time periods (Table 4). A significant *ln* [THg] versus age relationship was apparent in samples from both time periods, with an ANCOVA indicating no difference between the slopes of the time-dependent models (i.e., no age × time period interaction, $p = 0.92$), so the interaction term was removed from the model. The resulting ANCOVA ($R^2 = 0.50$, $F_{2,42} = 20.6$, $p < 0.001$) indicated a significant difference in the model intercepts ($p < 0.01$), with a higher age-specific *ln* [THg] in the 2008 sample compared to the 1978 sample (Fig. 2B). The relationship between *ln* [THg] and fork-length was not significant in either the 1978 or 2008 sample (Table 3).

Tikkoatokak Bay was sampled in both 1978 ($n = 25$) and 2007 ($n = 20$). There was no significant difference in mean age between the two time periods, but fork-lengths were greater in 1978 (Table 4). There was no significant difference in the mean or variance of *ln* [THg] between the two time periods (Table 4). Fish age was significantly related to *ln* [THg] during both time periods, with an ANCOVA ($R^2 = 0.25$, $F_{3,40} = 4.5$, $p < 0.01$) indicating similar slopes ($p = 0.48$) and intercepts ($p = 0.62$) of the time-dependent models (Fig. 2C). The relationship between *ln* [THg] and fork-length was significant only in the 2007 sample (Table 3).

Arctic charr were obtained from Voisey Bay in 1978 ($n = 25$) and 2007 ($n = 11$). The 1978 fish were older than those from 2007, but the fork-length distributions were similar between the two time periods (Table 4). Mean mercury concentrations did not differ between the two sampling periods, but the recent sample had a greater variance in *ln* [THg] (Table 4). Voisey Bay Arctic charr demonstrated no relationships between *ln* [THg] and age or fork-length during either time period (Table 3; Fig. 2D).

3.2. Non-anadromous Arctic charr

Individual fish [THg] ranged from 0.04 to 1.75 μg/g ww in 1977–78, and from 0.03 to 0.90 μg/g ww in 2007–09, with site means provided in Table 2. The linear mixed model estimated to explain mercury concentration using time period, given the random effect of sampling site, produced *ln* [THg] estimates of -1.72 ± 0.13 (mean ± standard error) or 0.18 μg/g ww for 1977–78, and -1.97 ± 0.13 or 0.14 μg/g ww for 2007–09, and the difference in mean [THg] between time periods was significant ($F_{1,243} = 3.8$, $p = 0.05$). When the model was estimated using fish age and fork-length as covariates, there were significant effects of fish age (linear mixed model; $F_{1,231} = 330.9$, $p < 0.001$) and fork-length ($F_{1,231} = 54.3$, $p < 0.001$) but no effect of sampling period ($F_{1,231} = 0.6$, $p = 0.46$) on non-anadromous Arctic charr mercury concentration.

Arctic charr ages ranged from 4 to 28 years in 1977–78 and from 1 to 30 years in 2007–09, with site means provided in Table 2. A linear mixed model produced mean age estimates of 11.7 ± 0.9 years (mean ± standard error) for the early time period and 10.5 ± 0.8 years for the recent time period, but the effect of time period on fish age was not significant ($F_{1,234} = 2.34$, $p = 0.13$). Measured fork-lengths ranged from 175 to 665 mm in the early sampling period and from 96 to 546 mm in the recent sampling period. A linear mixed model estimated fork-

Table 3
Pearson's correlation coefficients for the *ln* [THg] (μg/g ww) versus age (years), *ln* [THg] (μg/g ww) versus fork-length (mm), and age (years) versus fork-length (mm) relationships within each sample site and year. Significance of the correlation is indicated as: * = p < 0.05, ** = p < 0.01, *** = p < 0.001.

Life-history type	Sample site[a]	Year	n	*ln* [THg] vs. age[c]	*ln* [THg] vs. fork-length	Age vs. length[c]
Anadromous	Double Mer Lake	1977	12	NA	−0.22	NA
	Hebron Fiord[b]	1977	50	0.15	0.09	0.41**
	Hopedale Bay	1978	25	NA	0.07	NA
	Ikkudliayuk Fiord	1978	67	0.18	−0.13	0.55***
	Makkovik Bay	1978	18	0.40	0.43	0.38
	Nachvak Lake	1978	21	−0.13	−0.18	0.70***
	Okak Bay[b]	1978	25	0.57**	0.39	0.53**
	Ramah Bay	1978	25	0.07	−0.24	0.54**
	Sandhill River	1977	11	NA	0.16	NA
	Tikkoatokak Bay[b]	1978	25	0.40*	0.01	0.39
	Umiakovik Lake	1978	50	0.70***	0.66***	0.66***
	Voisey Bay[b]	1978	25	−0.03	−0.04	0.61**
	Anaktalak Bay	2007	20	0.51*	0.24	0.56**
	Hebron Fiord[b]	2009	21	0.67***	0.73***	0.73***
	Nain Bay	2008	20	0.63**	0.31	0.73***
	Okak Bay[b]	2008	20	0.80***	0.25	0.69***
	North Arm, Saglek	2007	22	0.89***	0.75***	0.83***
	Throat Bay	2007	17	0.19	0.14	0.81***
	Tikkoatokak Bay[b]	2007	20	0.53*	0.72***	0.60**
	Voisey Bay[b]	2007	11	0.10	0.30	0.74**
Non-anadromous	Esker Lake[b]	1978	26	0.65***	0.88***	0.76***
	Ikarut Lake	1978	10	0.59	0.91***	0.37
	Mistinippi Lake	1978	13	0.68*	0.60*	0.78**
	Saglek Lake	1978	17	0.53*	0.32	0.31
	Shapio Lake	1977	30	0.75***	0.85***	0.93***
	Tasialuk Lake[b]	1978	24	0.57***	0.17	0.60**
	Coady's Pond #2	2007	20	0.69***	0.68***	0.96***
	Esker Lake[b]	2008	24	0.81***	0.63**	0.88***
	Hebron Lake #2	2008	25	0.86***	0.81***	0.92***
	Tasialuk Lake[b]	2007	24	0.50*	−0.31	0.09
	Upper Nakvak Lake	2007	20	0.95***	0.88***	0.93***

[a] The following sample sites were omitted due to small sample size (n < 10 individuals captured): Anaktalik Lake, Napaktok Bay, Pack's Harbour, Cabot Lake, Komaktorvik Lake, Mistastin Lake, Nipishish Lake, St. Paul River, Shallow Lake.
[b] Sampled during both the early (1977–78) and recent (2007–09) time periods.
[c] NA = not analysed due to small sample size (age determined for n < 10 individuals).

lengths of 402 ± 15 mm for the early time period and 335 ± 18 mm for the recent time period, and indicated a significant reduction in the mean fork-length of sampled non-anadromous Arctic charr from 1977–78 to 2007–09 ($F_{1,242} = 12.9$, $p < 0.001$). Within a given sampling site and year, there was a significant positive correlation between *ln* [THg] and age in 10 of 11 samples (91%) with a sample size ≥10 (Table 3). There were significant positive correlations between [THg] and fork-length in 8 of 11 samples (73%) with a sample size ≥10 (Table 3). In most cases (73%), the correlation between mercury concentration and fish age was stronger than the correlation with fork-length (Table 3). Age and fork-length were significantly correlated in 8 of 11 samples with a sample size ≥10 (Table 3).

Non-anadromous Arctic charr were captured from Esker Lake in 1978 (n = 26) and 2008 (n = 24). Mean *ln* [THg], fork-length, and age were higher in the early sampling period, though the difference for age was not statistically significant (Table 4). Mercury concentration

Table 4
Comparison of Arctic charr *ln* [THg] (μg/g ww), age (years), and fork-length (mm) between 1977–78 and 2007–09 at locations sampled during both time periods. The life-history type of Arctic charr at each location is given as anadromous (A) or non-anadromous (N). Tests indicating significantly different means or variances between the two sample periods are shown in bold.

Sample site	Life-history	Variable	Comparison of means	Comparison of variances
Hebron Fiord	A	*ln* [THg]	**Welch t-test; t = −4.91, df = 68.0, p < 0.001**	**F-test; $F_{49,20} = 4.54$, p < 0.001**
		Age	t-test; t = 0.28, df = 66, p = 0.78	Levene's test; $F_{1,66} = 1.05$, p = 0.31
		Fork-length	t-test; t = −0.30, df = 69, p = 0.77	Levene's test; $F_{1,69} = 1.28$, p = 0.26
Okak Bay	A	*ln* [THg]	t-test; t = −1.23, df = 43, p = 0.23	F-test; $F_{24,19} = 0.85$, p = 0.71
		Age	t-test; t = 1.58, df = 43, p = 0.12	Levene's test; $F_{1,43} = 0.46$, p = 0.50
		Fork-length	t-test; t = 1.23, df = 43, p = 0.23	Levene's test; $F_{1,43} = 3.53$, p = 0.07
Tikkoatokak Bay	A	*ln* [THg]	t-test; t = 1.54, df = 43, p = 0.13	F-test; $F_{24,19} = 0.97$, p = 0.93
		Age	Welch t-test; t = 0.95, df = 23.7, p = 0.35	**Levene's test; $F_{1,42} = 7.09$, p = 0.01**
		Fork-length	**t-test; t = 4.85, df = 43, p < 0.001**	Levene's test; $F_{1,43} = 0.45$, p = 0.51
Voisey Bay	A	*ln* [THg]	Welch t-test; t = −1.35, df = 11.6, p = 0.20	**F-test; $F_{24,10} = 0.18$, p < 0.001**
		Age	**Wilcoxon test; W = 184.5, p = 0.03**	Levene's test; $F_{1,32} = 1.52$, p = 0.23
		Fork-length	t-test; t = 1.18, df = 33, p = 0.25	Levene's test; $F_{1,33} = 2.19$, p = 0.15
Esker Lake	N	*ln* [THg]	**t-test; t = 5.05, df = 48, p < 0.001**	F-test; $F_{25,23} = 0.61$, p = 0.23
		Age	t-test; t = 1.84, df = 47, p = 0.07	Levene's test; $F_{1,47} = 0.81$, p = 0.37
		Fork-length	**t-test; t = 5.53, df = 48, p < 0.001**	Levene's test; $F_{1,48} = 0.04$, p = 0.84
Tasialuk Lake	N	*ln* [THg]	**t-test; t = −10.63, df = 46, p < 0.001**	F-test; $F_{23,23} = 1.66$, p = 0.23
		Age	t-test; t = −1.04, df = 46, p = 0.31	Levene's test; $F_{1,46} = 1.00$, p = 0.32
		Fork-length	**t-test; t = −5.4, df = 46, p < 0.001**	Levene's test; $F_{1,46} = 0.69$, p = 0.41

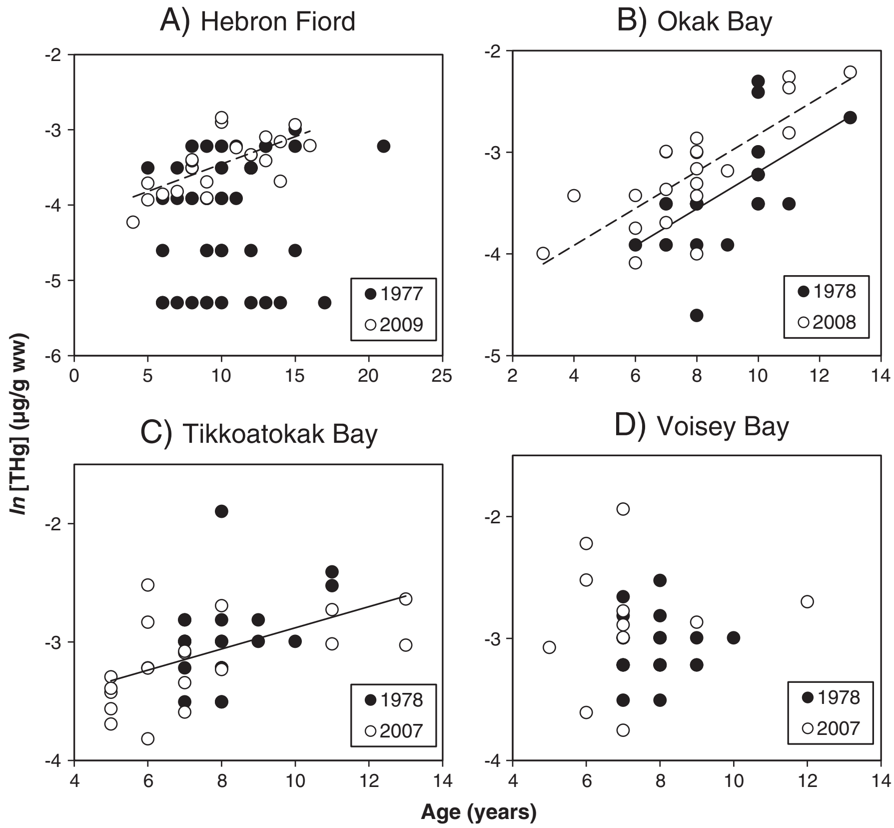

Fig. 2. Relationships between *ln* [THg] (μg/g ww) and age (years) in anadromous Arctic charr captured from common sampling sites in 1977–78 (filled circles) and 2007–09 (open circles). [A] Hebron Fiord: a significant relationship was observed only in the 2009 sample (dashed line). [B] Okak Bay: there was no significant difference in the slopes of the group-dependent models, thus common-slope models were estimated for 1978 (solid line) and 2008 (dashed line). [C] Tikkoatokak Bay: there was no significant difference between the two time-dependent models, thus a single model was fitted using the data from both time periods (solid line). [D] Voisey Bay: no significant relationship was observed in either time period.

was significantly positively related to age and fork-length during both time periods (Table 3). The *ln* [THg] versus age relationships for the two time periods had the same slope (no interaction between age and time period, $p = 0.54$), therefore the interaction term was removed from the ANCOVA model. The resulting model (ANCOVA; $R^2 = 0.71$, $F_{2,46} = 56.5$, $p < 0.001$) indicated significantly different intercepts between the 1978 and 2008 models ($p < 0.001$), with age-specific *ln* [THg] in the early sample exceeding the recent sample (Fig. 3A). ANCOVA ($R^2 = 0.71$, $F_{3,46} = 36.7$, $p < 0.001$) identified no differences in the slopes ($p = 0.67$) or intercepts ($p = 0.98$) of the *ln* [THg] versus fork-length relationships between the two time periods (Fig. 3B).

Tasialuk Lake was sampled in 1978 ($n = 24$) and 2007 ($n = 24$). There was no significant difference in mean age between the two time periods, but the recent sample had significantly higher mean fork-length and *ln* [THg] than the early sample (Table 4). Significant positive relationships between *ln* [THg] and age were apparent in both time periods, but relationships with fork-length were not significant (Table 3; Fig. 3C and D). ANCOVA indicated no difference in the slopes of the *ln* [THg] versus age relationship between the two time periods ($p = 0.26$), therefore the interaction term was removed and the resulting model ($R^2 = 0.79$, $F_{2,45} = 84.2$, $p < 0.001$) demonstrated a significant difference in the intercepts ($p < 0.001$). Age-specific mercury concentrations in 2007 were higher than in 1978 (Fig. 3C).

3.3. Comparison of the two life-history types

The linear mixed model used to explain age as a function of life-history type, given the random effect of sample site, produced mean age estimates of 9.0 ± 0.6 years (mean ± standard error) for anadromous and 11.8 ± 1.0 years for non-anadromous Arctic charr captured in 1977–78. During the early sampling period, the mean age of anadromous Arctic charr was significantly lower than the mean age of non-anadromous conspecifics (linear mixed model; $F_{1,25} = 7.5$, $p = 0.01$; Fig. 4). The same pattern was observed in the 2007–09 samples (Fig. 4), where mean age estimates were 8.0 ± 0.7 years for anadromous and 10.5 ± 1.1 years for non-anadromous fish, and the difference between the two life-history types was significant (linear mixed model; $F_{1,11} = 5.4$, $p = 0.04$). When the data from both time periods were considered together, the mean age of captured Arctic charr was significantly higher in 1977–78 than in 2007–09 ($F_{1,696} = 6.1$, $p = 0.01$), and the decline was statistically similar in anadromous and non-anadromous fish (no significant life-history type × time period interaction; $p = 0.45$).

For fish captured during the early sampling period, the estimated mean fork-length was significantly higher (linear mixed model; $F_{1,25} = 5.8$, $p = 0.02$; Fig. 5) in anadromous Arctic charr (467 ± 18 mm) than in non-anadromous conspecifics (401 ± 28 mm). Similarly, the estimated mean fork-length of anadromous fish (408 ± 24 mm) exceeded that of non-anadromous Arctic charr (332 ± 39 mm) during 2007–09, but the difference was not statistically significant in the later sampling period (linear mixed model; $F_{1,11} = 3.9$, $p = 0.07$; Fig. 5). When the data from both time periods were considered together, the mean fork-length of captured Arctic charr significantly decreased from 1977–78 to 2007–09 ($F_{1,745} = 19.0$, $p < 0.001$), and the pattern was similar in anadromous and non-anadromous fish (no significant life-history type × time period interaction; $p = 0.25$).

For samples from 1977–78, the estimated mean *ln* [THg] was −3.45 ± 0.15 or 0.03 μg/g ww in anadromous and −1.70 ± 0.23 or 0.18 μg/g ww in non-anadromous Arctic charr, and there was a significant effect of life-history type on mercury concentration (linear mixed model; $F_{1,25} = 56.2$, $p < 0.001$; Figs. 4 and 5). The pattern was similar in the 2007–09 samples (Figs. 4 and 5), with the estimated mean *ln*

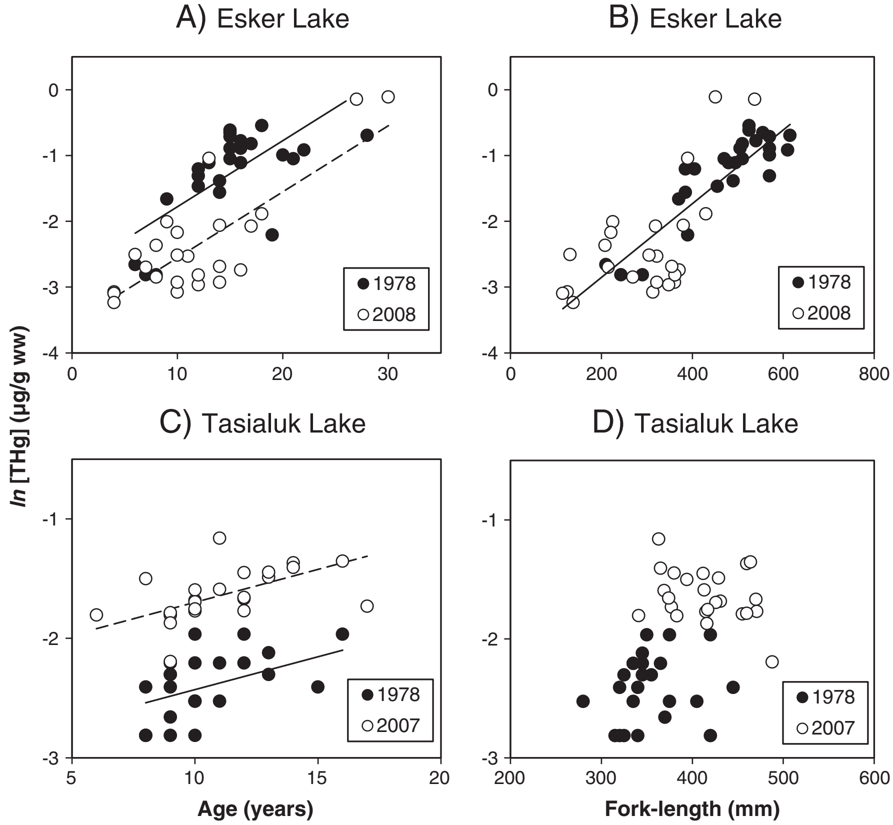

Fig. 3. Relationships between *ln* [THg] (μg/g ww) and covariates in non-anadromous Arctic charr captured from common sampling sites in 1978 (filled circles) and 2007–08 (open circles). [A] Esker Lake, *ln* [THg] (μg/g ww) versus age (years): there was no significant difference in the slopes of the group-dependent models, thus common-slope models were estimated for 1978 (solid line) and 2008 (dashed line). [B] Esker Lake, *ln* [THg] (μg/g ww) versus fork-length (mm): there was no significant difference between the two time-dependent models, thus a single model was fitted using the data from both time periods (solid line). [C] Tasialuk Lake, *ln* [THg] (μg/g ww) versus age (years): There was no significant difference in the slopes of the group-dependent models, thus common-slope models were estimated for 1978 (solid line) and 2008 (dashed line). [D] Tasialuk Lake, *ln* [THg] (μg/g ww) versus fork-length (mm): no significant relationship was observed in either time period.

[THg] for anadromous Arctic charr (-3.34 ± 0.14 or 0.04 μg/g ww) was significantly lower (linear mixed model; $F_{1,11} = 28.6$, $p < 0.001$) than for non-anadromous fish (-2.11 ± 0.23 or 0.12 μg/g ww). When the data from both time periods were considered together, there was no significant change in mean *ln* [THg] between 1977–78 and 2007–09 ($p = 0.19$).

3.4. Temperature trends

Large fluctuations in mean annual temperature occurred from year to year, and there was an anomalous cold period in Labrador during the late 1980s and early 1990s (Fig. 6). Overall, a significant warming trend was observed at all four weather stations between 1970 and 2010 (linear regressions, all $p < 0.01$). Slope estimates ranged from 0.05 °C/year (Cartwright) to 0.10 °C/year (Hopedale), indicating an increase in mean annual temperature of 1.6 °C to 2.9 °C over a 30 year period (e.g., from 1978 to 2008).

4. Discussion

Mercury concentrations were compared between 1977–78 and 2007–09 in Arctic charr from a variety of locations in Labrador, Canada. After

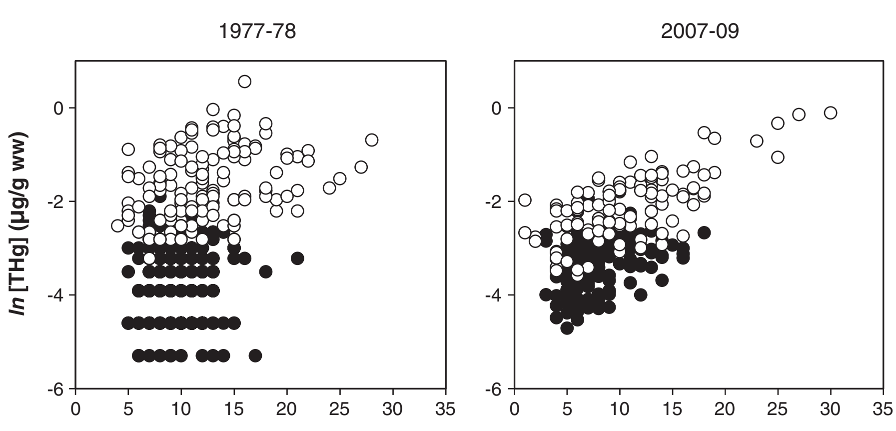

Fig. 4. *ln* [THg] (μg/g ww) versus age (years) in anadromous (filled circles) and non-anadromous (open circles) Arctic charr captured from 1977–78 and 2007–09. Data are included from all sampling locations, and points represent individual fish.

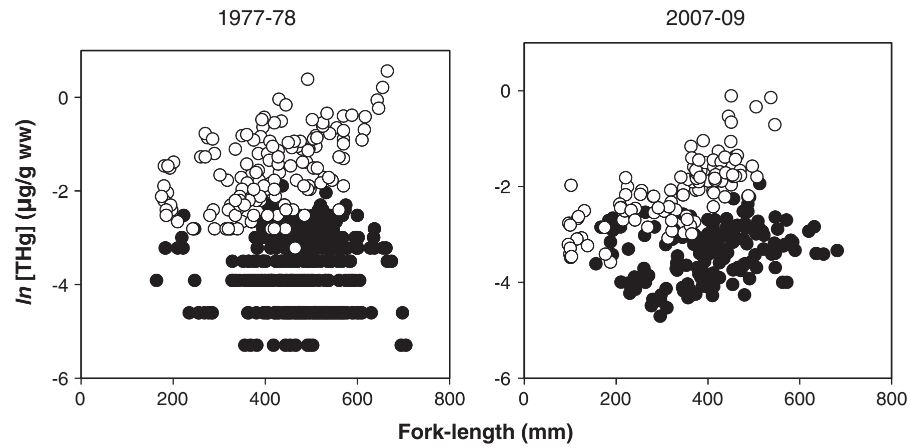

Fig. 5. *ln* [THg] (μg/g ww) versus fork-length (mm) in anadromous (filled circles) and non-anadromous (open circles) Arctic charr captured from 1977–78 and 2007–09. Data are included from all sampling locations, and points represent individual fish.

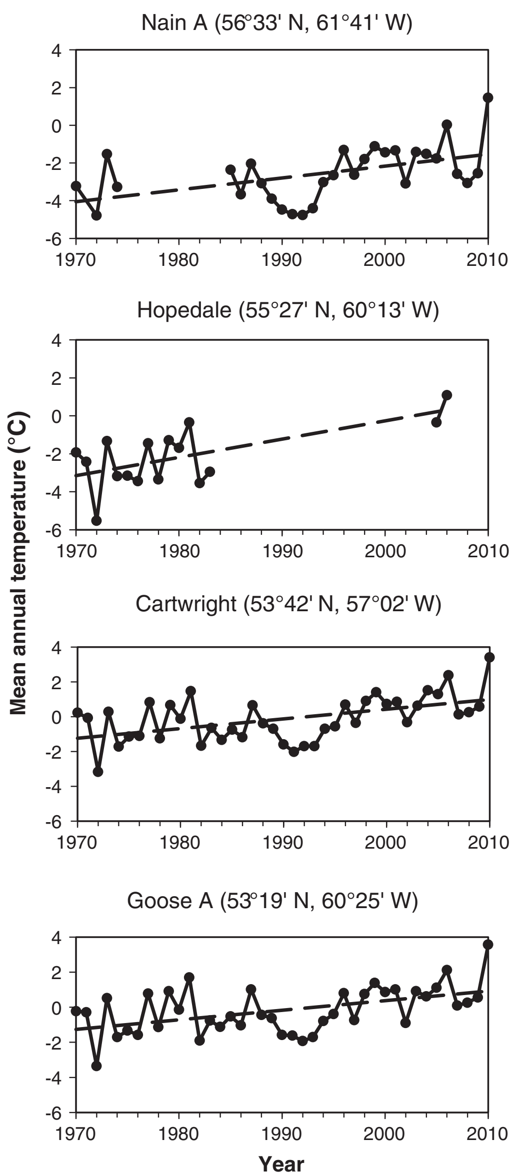

Fig. 6. Mean annual temperatures (°C) from 1970 to 2010 at four weather stations in Labrador, Canada. Data were obtained from Environment Canada National Climate Data and Information Archive (www.climate.weatheroffice.gc.ca).

correcting for differences in age and fork-length, there was insufficient evidence to suggest that there has been a widespread change in [THg] of anadromous or non-anadromous Arctic charr over the past thirty years. Mercury concentrations were positively related to age and fork-length in many but not all samples, and the relationships were usually stronger with age and in non-anadromous populations. The mean age and fork-length of captured fish declined from 1977–78 to 2007–09, and the trends were similar in Arctic charr of both life-history types. Anadromous Arctic charr had lower mean [THg], lower mean age, and greater mean fork-length than non-anadromous conspecifics. Throughout the region, the mean annual temperature increased by 1.6 °C to 2.9 °C between the two sampling periods, without a corresponding change in mean [THg].

The lack of a widespread change in Arctic charr [THg] between 1977–78 and 2007–09 supports the general lack of consistent mercury temporal trends observed in studied Arctic biota over the past thirty years (Rigét et al., 2011). Our results imply that mercury concentrations in Labrador lacustrine and marine fish habitats and prey resources have remained constant over the past three decades (Braune et al., 2011). Evidence from elsewhere suggests that the global atmospheric mercury pool, and therefore the rate of mercury deposition, has been relatively stable or declining in recent decades. Global anthropogenic mercury emissions to the atmosphere were in the range of 3500 ton/year during the late 1970s and early 1980s, but decreased during the 1980s to reach relatively stable levels of approximately 1900 to 2200 ton/year from 1990 to 2005 (Pacyna et al., 2006, 2010). Consequently, direct measurements of atmospheric mercury have indicated steady or declining concentrations since the 1970s at Canadian temperate and Arctic sites (Li et al., 2009; Steffen et al., 2005; Temme et al., 2007) and globally (Lindberg et al., 2007). Evidence from the interstitial air of perennial snowpack (Faïn et al., 2009), glacial ice (Schuster et al., 2002), peat bogs (Shotyk et al., 2005), and mercury concentrations in precipitation (Temme et al., 2007) also indicates consistent or diminishing mercury deposition across the northern hemisphere during recent decades. Recent declines in mercury flux to lake sediments were evident in only 18% of Arctic, 25% of sub-Arctic, and 28% of midlatitude lake cores (Muir et al., 2009). However, recent increasing fluxes to northern lake sediments may reflect mercury retention and slow release from lake catchments (Munthe et al., 2007), and/or climate change related increases in mercury inputs (e.g., permafrost degradation and erosion) or algal scavenging (Kirk et al., 2011; Outridge et al., 2007), rather than continuing increases in atmospheric deposition.

Fish mercury concentrations are positively related to atmospheric mercury deposition (Hammerschmidt and Fitzgerald, 2006). However, the response of fish [THg] to changing mercury inputs is site-specific,

depending on a variety of physical, chemical, and biological factors (Munthe et al., 2007). Consequently, Arctic charr [THg] increased at Hebron Fiord and Tasialuk Lake, and age-specific [THg] increased at Okak Bay, despite the lack of a widespread change in fish [THg] throughout Labrador. At the same time, age-specific [THg] decreased at Esker Lake, but there was no corresponding reduction in length-specific mercury concentration. Our results support previous conclusions that regional characteristics (e.g., mean annual temperature, atmospheric mercury deposition) are less important than site-specific factors (e.g., population age and size distribution, food web structure, fish diet composition) in determining fish [THg] (e.g., Rose et al., 1999; van der Velden et al., 2013a).

A decrease in the mean age and size of Arctic charr captured from the Labrador coast since the 1970s has been previously documented. Dempson et al. (2008) noted a reduction in the mean age of anadromous Arctic charr captured from the Nain and Okak Bay regions throughout the 1980s and 1990s, and a decrease in the mean weight of Nain, Okak, and Voisey Bay fish from 1977 to 1997. All stocks showed recent increases in mean age and weight since minimum values in the late 1990s. The observed decline in the mean size and age of Labrador Arctic charr could be related to differences in environmental conditions, prey type and availability and/or commercial fishing pressure (Dempson et al., 2002, 2008; Michaud et al., 2010). Alternatively, the apparent differences in fish age and size between 1977–78 and 2007–09 may be an artefact of different sampling techniques, rather than a change in population characteristics. For example, gillnets of unknown mesh sizes were used to capture non-anadromous fish in the early sampling period, and the efficiency of fish capture is known to vary with mesh size such that smaller mesh gillnets more effectively capture smaller fish (e.g., Bromaghin, 2005). The gillnet mesh sizes used to capture anadromous Arctic charr have remained constant since the 1970s (Dempson et al., 2008), however, a difference in the timing of the anadromous Arctic charr fishery may be contributing to the observed difference in fish size distribution (Dempson, 1995). In spite of the decreased mean age and length observed between the two sampling periods, there was no reduction in the mercury concentration of anadromous Arctic charr. One possible reason for this discrepancy is the weaker link between [THg] and age or length in anadromous individuals compared to non-anadromous fish (Table 3; van der Velden et al., 2013a).

The lack of an overall trend in fish [THg] observed, despite a warming temperature trend that favours increased mercury methylation, suggests that regional changes in climate-driven factors (e.g., mean annual temperature, primary productivity, extent and duration of ice cover) have thus far had limited impacts on mercury exposure in Labrador anadromous or non-anadromous Arctic charr. Similarly, previous studies have identified no relationship between site latitude and [THg] in anadromous or non-anadromous Arctic charr from Canada (56–82°N, Gantner et al., 2010; 49–81°N, van der Velden et al., 2013a). In contrast, Carrie et al. (2010) found increasing mercury concentrations between 1985 and 2008 in burbot (*Lota lota*) from the Mackenzie River, Canada despite consistent atmospheric deposition, and concluded that climate-driven environmental processes were important for explaining the trend in fish [THg]. The difference in results between this study and that of Carrie et al. (2010) might arise because the impact of climate warming on Labrador aquatic ecosystems has been limited in comparison to the western Canadian Arctic (Smol et al., 2005). Alternatively, the trend in burbot [THg] may relate to the benthic nature of the species ecology, providing a more direct link to climate change related increases in sediment mercury concentrations (Carrie et al., 2010) and/or mercury methylation occurring in Arctic lake sediments (Hammerschmidt et al., 2006). Together these findings provide further evidence that climate change effects on fish [THg] are site-specific, depending on local factors such as mercury inputs from lake catchments, mercury methylation capacity of the system, and aquatic food web structure (Macdonald et al., 2005).

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